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Human health screening and public health significance of contaminants of emerging concern detected in public water supplies

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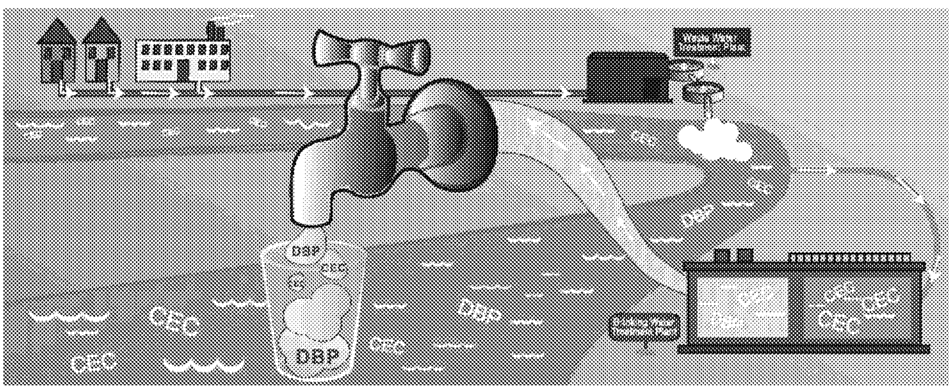
HIGHLIGHTS

- Water samples from 25 drinking water treatment plants were collected in 2010-2012.
- The water samples were analyzed for 247 contaminants.
- The health significance of the contaminants in the treated water was assessed.
- The measure of the health significance was the margin of exposure.
- This analysis shows only a small number of contaminants raise a health concern.

GRAPHICAL ABSTRACT



Abbreviations: ACToR, Aggregated Computational Toxicology Resource; ADI, Acceptable Daily Intake; ATC, Anatomical Therapeutic Chemical; ATSDR, Agency for Toxic Substances and Disease Registry; AWI, anthropogenic waste indicator; BMDL, Lower 95% Confidence Limit of the Benchmark Dose; CECs, contaminants of emerging concern; DWEL, Drinking Water Equivalent Level; DWTP, drinking water treatment plant; E1, estrone; E2, 17β-estradiol; E3, estriol; EE2, 17α-ethinyl estradiol; HHCB, hexahydrohexamethyl cyclopentabenzopyran; IRIS, Integrated Risk Information System; LCMRL, lowest concentration minimum reporting limit; LOAEL, Lowest Observed Adverse Effect Level; MCL, Maximum Contaminant Level; MOE, Margin of Exposure; MRTD, Maximum Recommended Therapeutic Dose; NOAEL, No Observed Adverse Effect Level; PFAs, perfluorinated acids; PFOA, perfluorooctanoic acid; PFOS, perfluorooctanesulfonic acid; PPRTV, Provisional Peer Reviewed Toxicity Value; PFSs, perfluorinated sulfonic acids; RfD, Reference Dose; RSL, Relative Source Contribution; UF, uncertainty factor; USEPA, U. S. Environmental Protection Agency; USGS, U. S. Geological Survey; WHO, World Health Organization.

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ABSTRACT

The source water and treated drinking water from twenty five drinking water treatment plants (DWTPs) across the United States were sampled in 2010–2012. Samples were analyzed for 247 contaminants using 15 chemical and microbiological methods. Most of these contaminants are not regulated currently either in drinking water or in discharges to ambient water by the U. S. Environmental Protection Agency (USEPA) or other U.S. regulatory agencies. This analysis shows that there is little public health concern for most of the contaminants detected in treated water from the 25 DWTPs participating in this study. For vanadium, the calculated Margin of Exposure (MOE) was less than the screening MOE in two DWTPs. For silicon, the calculated MOE was less than the screening MOE in one DWTP. Additional study, for example a national survey may be needed to determine the number of people ingesting vanadium and silicon above a level of concern. In addition, the concentrations of lithium found in treated water from several DWTPs are within the range previous research has suggested to have a human health effect. Additional investigation of this issue is necessary. Finally, new toxicological data suggest that exposure to manganese at levels in public water supplies may present a public health concern which will require a robust assessment of this information.

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1. Introduction

Water is a necessary component of life. Yet there is a clear recognition that current human use of available fresh water is not sustainable. The presence in drinking water of chemicals derived from human inputs into source water is of increasing public concern with regard to both sustainability and public health. Ideally, the water one consumes should be free of harmful chemical and microbial contaminants. The Safe Drinking Water Act defines "contaminant" as any physical, chemical, biological or radiological substance or matter in water. However, source waters used to produce drinking water often contain both anthropogenic and naturally occurring contaminants. The anthropogenic contaminant load results from the complex interplay of increases in population growth, chemical, consumer product, and pharmaceutical usage per consumer and the number of times a particular unit of water is re-used as it moves through the watershed. While it is technologically possible to remove most contaminants to levels below analytical detection limits, the implementation of the treatment technology required to do so could make the water prohibitively expensive. In addition, the presence of some minerals (e.g., magnesium sulfate, potassium chloride, sodium chloride, calcium chloride, magnesium chloride, and potassium bicarbonate) generally improves the taste of drinking water and their presence is considered beneficial. The goal of the drinking water treatment plant (DWTP) is to provide safe drinking water for humans, which is to reduce the concentrations such that any remaining contaminants do not pose an unacceptable human health risk.

This paper is one of a series of papers (Glassmeyer et al., 2016; Furlong et al., 2016; Conley et al., 2016) describing a comprehensive study on the presence, concentrations, and persistence of chemical and microbial contaminants of emerging concern (CECs) in source and treated drinking waters of the United States. This was a joint effort of the U.S. Environmental Protection Agency and the U.S. Geological Survey. A primary goal of this study was to provide information for assessing the potential for human exposure to CECs via drinking water. A secondary goal was to estimate removal efficiency of CECs from source waters by currently used drinking water treatment processes under typical DWTP operating conditions, and thus identify possible compounds or organisms that may be amenable to enhanced reduction or removal. The objective of the analysis reported here is to apply health screening values to the contaminants detected in treated drinking water to assess the potential of the detected contaminants to pose a human health risk from long-term exposure.

2. Experimental (materials and methods)

In 2010–2012, USEPA arranged for the collection of paired samples of source and treated water from twenty-five DWTP across the United States (Supporting Information Table 1) (Glassmeyer et al., 2016). A goal of this study was to better determine the upper boundary of CEC concentrations, rather than provide a nationwide average, so DWTP selection was skewed towards sample locations with known wastewater outfalls in the source water. Candidate locations were selected based on integrated wastewater and drinking water reports (Swayne et al. 1980), locations with and without existing pharmaceutical concentration data (Associated Press), nomination by USEPA and USGS regional personnel, and DWTP self-nomination. Sites were chosen to maximize geographic range, diversity in disinfectant type used in the treatment process, and drinking water plant production volume. Participation in the study was voluntary.

These water samples were analyzed for 247 chemical and microbial contaminants using 15 chemical and microbial methods. The complete description of the analytical methods, the detection limits, and the concentrations detected in source and treated drinking water for the chemical contaminants are presented elsewhere (Glassmeyer et al., 2016; Furlong et al., 2016; Conley et al., 2016). An overview of the analytical methods is provided in Supporting Information Table 2. The focus of the analysis presented here is on contaminants of emerging concern (CECs) detected in treated drinking water in comparison to human health information from long-term exposure to the contaminant. Accordingly, chemicals with existing Maximum Contaminant Levels (MCLs) for drinking water (U.S. Environmental Protection Agency) were excluded (antimony, arsenic, atrazine, barium, bromate, cadmium, chlorite, chromium, copper, fluoride, lead, nitrate, nitrite, selenium, and uranium). Also excluded from this analysis are select chemicals that are essential nutrients (calcium, chloride, magnesium, phosphorus, potassium, and sodium) and chemicals with reference values based on aesthetic effects (taste and odor) (ammonia and sulfate) rather than adverse health effects. Although iron and zinc are essential nutrients, they are included in this analysis because there is concern for adverse health effects at elevated exposure (U.S. Environmental Protection Agency; U.S. Environmental Protection Agency Integrated Risk Information System (IRIS)). Manganese is included because new information suggests the potential for adverse developmental neurological effects in the range of exposures (100 to 1000 μg/L) often found in drinking water supplies (Ljung and Vahter, 2007; Menezes-Filho et al., 2009; Bouchard et al., 2011; Oulhote et al., 2014).

A variety of perfluorinated chemicals were detected in the treated drinking water of every DWTP. The list of these analytes is in Supporting Information Table 4. However, an analysis of the human health significance from exposure to these chemicals is not presented in this publication. The analysis of the human health significance of exposure to PFSs and PFAs will be reported in a future publication.

Information on health effects for chemicals (expressed as mg/kg body weight per day) was obtained from a variety of sources, including the USEPA Integrated Risk Information System (IRIS) data base (U.S. Environmental Protection Agency Integrated Risk Information System (IRIS)), the USEPA Office of Water Provisional Health Advisories, the USEPA Superfund Provisional Peer Reviewed Toxicity Value (PPRTV) documents (U.S. Environmental Protection Agency), the Agency for Toxic Substances and Disease Registry (ATSDR) Toxicological Profiles (Agency for Toxic Substances and Disease Registry (ATSDR), 2005), the USEPA Office of Pesticide Programs Registration Eligibility Decision documents (U.S. Environmental Protection Agency), the World Health Organization Joint Expert Commission on Food Additives Acceptable Daily Intake (ADI) documents (World Health Organization), the National Sanitation Foundation documents (National Sanitation Foundation International), and USEPA's Aggregated Computational Toxicology Resource (ACToR) data base (U.S. Environmental Protection Agency).

The analysis reported here followed USEPA's risk assessment methodology as described in IRIS (U.S. Environmental Protection Agency). Useful background documents found in IRIS under "Guidance and Tools" include "Reference Dose (RfD): Description and Use in Health Risk Assessments" and "A Review of the Reference Dose and Reference Concentration Processes." The health assessment document for each substance provides information of the toxicity benchmark for the adverse health effect for the substance, that is, the No Observed Adverse Effect Level (NOAEL), the Lower 95% Confidence Limit of the Benchmark Dose (BMDL), or the Lowest Observed Adverse Effect Level (LOAEL) from a long-term toxicity study. Each health assessment document also provides information on the uncertainty factors (UF) used in the assessment. The types of UFs can include uncertainty in extrapolating from a laboratory animal to a human (UF_A), uncertainty in extrapolating to the general human population (UF_H), uncertainty in extrapolating from a sub-chronic to a chronic exposure (UF_S), uncertainty in extrapolating from a LOAEL to a NOAEL (UF_L), and uncertainty due to an incomplete data base (UF_D). The total UF used in the assessment is chemical specific and depends on the quality and quantity of the toxicological data available.

The margin of exposure (MOE) was used as a screening tool to assess whether or not exposure might present a significant public health concern from long-term exposure to the contaminants detected in treated water. The MOE is a ratio of a toxicity benchmark and an exposure dose. The Drinking Water Equivalent Level (DWEL, mg/L) was calculated from the NOAEL, the BMDL, or the LOAEL and the standard drinking water scenario (80 kg person drinking 2.4 L of water per day).

$$\text{DWEL (mg/L)} = \frac{(\text{NOAEL, BDML or LOAEL})\ \text{mg/kg} - \text{day} \times 80\ \text{kg}}{2.4\ \text{L water/day}}$$

The MOE for each chemical was then calculated by dividing the DWEL by the concentration detected in the treated drinking water and rounded to two significant digits.

$$\text{MOE} = \frac{\text{DWEL}}{\text{Measured Drinking Water Concentration}}$$

For purposes of this analysis, the assumption is that drinking water provides 100% of the contaminant source contribution. In the absence of contaminant specific data on exposure from other media, a data derived Relative Source Contribution (RSC) cannot be calculated. If a default 20% RSC were to be applied, the MOE values would be lower. The calculated MOE was then compared to the MOE screening value. Considerations for selecting the screening MOE for each chemical include the quality and quantity of the toxicological data available for a particular contaminant as reflected in the total UF used in the human health assessment. The screening value for the MOE for the chemicals was assigned as equal to the total uncertainty factor (UF) used in the human health assessment for the particular contaminant (U.S. Environmental Protection Agency; Agency for Toxic Substances and Disease Registry (ATSDR), 2005; U.S. Environmental Protection Agency; World Health Organization; National Sanitation Foundation International; U.S. Environmental Protection Agency). Silicon and hexahydrohexamethyl cyclopentabenzopyran (HHCB also known as galaxolide) do not have conventional human health assessments. For these chemicals the MOE screening value of 3000 was used as this is the maximum total UF allowed by the IRIS Program for a file that is posted on the data base (U.S. Environmental Protection Agency).

Because of the lack of available toxicity data for the majority of pharmaceuticals, the Maximum Recommended Therapeutic Dose (MRTD) was used to calculate the MOE. The MRTD is an estimated upper dose limit beyond which a drug's efficacy is not increased and/or undesirable adverse effects begin to outweigh beneficial effects recommended by the Food and Drug Administration to treat targeted patient populations for specific conditions (Contrera et al., 2004) and are clearly effect levels in the targeted patient population. These values are readily accessible via the Drugs.com internet database (Drugs.com, 2016). It should be noted that these values are developed for the targeted patient population and not for the general population, which includes potentially sensitive populations such as infants, pregnant women, and the immuno-compromised (Matthews et al., 2004). For all pharmaceuticals where the MRTD was used to calculate the DWEL, the MOE screening value of 3000 was used to be consistent with the process of deriving a Health Reference Level from the MRTD in the Contaminant Candidate List process (U. S. Environmental Protection Agency).

When the calculated MOE is larger than the selected MOE screening value, it is generally considered that exposure to that contaminant has little, if any, public health significance. In contrast, if the calculated MOE is smaller than the selected MOE screening value, then further research may be necessary to decide whether controls to limit exposure are warranted.

3. Results and discussion

Forty-one CECs were detected one or more times in the treated drinking waters at concentrations greater than their respective lowest concentration minimum reporting limit (LCMRL) in this study. MOEs were calculated for twenty-six analytes (three pharmaceuticals lack an adequate toxicity value and the 12 PFCs will be reported separately).

3.1. Assessment of chemicals detected in elemental analytes method

The calculated MOE reported in Table 1 is for the water system that had the highest detected level of the particular analyte in the treated drinking water (Glassmeyer et al., 2016). Additional details are in Supporting Information Table 3.

Vanadium was detected in treated drinking water in only four DWTPs (5, 23, 25, and 28). However, the calculated MOE for vanadium was less than the screening MOE of 3000 in treatment plants 5 (1500) and 25 (1600) and close to the screening MOE in treatment plant 23 (3200). Thus there is some public health concern for exposure to vanadium from drinking water and further research is necessary to decide whether controls to limit exposure to vanadium are warranted to increase public health protection.

Although the MOE for exposure to manganese is greater than the screening level MOE of 3, there is some potential public health concern from exposure to manganese from drinking water because there is evidence from new toxicological studies (Ljung and Vahter, 2007; Menezes-Filho et al., 2009; Bouchard et al., 2011; Oulhote et al., 2014) that the NOAEL in the general adult population could be an effect level for developmental neurotoxicity. However, a systematic review of the available data on manganese has not been conducted and a new NOAEL or LOAEL is not available.

Silicon was detected in the treated water from every DWTP. The calculated MOE for silicon was less than the screening MOE of 3000 in only one DWTP. However, 14 DWTPs had a calculated MOE between

3000 and 10,000 as indicated in Supporting Information Table 3. As there is uncertainty in the selected screening MOE due to the poor quality of the toxicity data base for silicon, it would be helpful if additional toxicity data were collected.

3.2. Assessment of anthropogenic waste indicators

USGS Analytical Method 1433 covers a wide variety of chemicals commonly present in wastewater, such as detergent metabolites, fragrances and pesticides, which we collectively refer to as anthropogenic waste indicators (AWIs). The MOE reported in Table 2 is for the water system that had the highest detected level of the particular analyte in the treated drinking water (Glassmeyer et al., 2016). Additional details are in Supporting Information Table 3. All of the calculated MOEs are more than 3 million. These results suggest that exposure to these compounds from drinking water is not likely to pose a public health concern.

3.3. Assessment of pharmaceuticals

Some general risk-related conventions to consider when evaluating pharmaceuticals as drinking water contaminants are: 1) for pharmaceuticals, risks of adverse effects are often tolerable in relation to therapeutic benefits whereas, for drinking water contaminants, adverse effects resulting from exposure are undesirable and would be expected to trigger remedial action to reduce exposure; 2) for pharmaceuticals, therapeutic pharmacological effects are expected under conditions of use, whereas for drinking water contaminants, pharmacological effects are to be avoided; and 3) pharmaceuticals are approved and intended for specific patient populations, whereas acceptable drinking water contaminant levels must be considered harmless to the general population (World Health Organization, 2011). Therefore, the Maximum Recommended Therapeutic Dose (MRTD) is not an ideal measure to be used when assessing pharmaceuticals as drinking water contaminants. However, in the absence of readily available toxicological data, the MRTD is considered the best available data for the purposes of this analysis. The approach used in this assessment is to consider the MRTD as a LOAEL in the calculation of the MOE and to use a screening MOE of 3000 when the MRTD was used to derive the DWEL.

Of the 118 pharmaceuticals included in this study, 41 were detected in at least one source water sample. The pharmaceuticals that were detected represent varied modes of action and drug types and are listed in Supporting Information Table 5 by drug type and World Health Organization (WHO) Anatomical Therapeutic Chemical (ATC) Classification System code. Fluconzole and diphenhydramine appear in two different WHO categories, while ibuprofen and lidocaine appear in three different WHO categories. Pharmaceuticals detected in source water can be categorized into 8 ATC categories: alimentary tract and metabolism, cardiovascular system, dermatologicals, genitourinary system and sex hormones, anti-infectives for systemic use, musculo-skeletal system, nervous system, and respiratory system (Supporting Information Table 5). However, after the drinking water treatment process, this number diminishes to 5. No pharmaceuticals from 3 ATC categories (alimentary tract and metabolism, dermatologicals or musculoskeletal system) were detected in treated drinking water.

Of the 23 pharmaceuticals detected in treated drinking water, only 13 were detected at concentrations greater than their respective LCMRL or an alternative minimum reporting limit (clofibric acid, lithium, and pseudoephedrine) (Furlong et al., 2016). MRTDs were not located for three of these pharmaceuticals (cotinine, sulfamethoxazole, and verapamil). The MOEs for the remaining nine pharmaceuticals were calculated and are listed in Table 3. Progesterone is discussed in Section 3.4, assessment of hormonally active environmental contaminants. The MOE reported in Table 3 is for the water system that had the highest detected level of the particular pharmaceutical in the treated drinking water (Furlong et al., 2016). Additional details are in Supporting Information Table 3. The screening MOE for all pharmaceuticals where the DWEL was calculated from the MRTD was 3000. There is a health assessment for lithium based on a NOAEL (U.S. Environmental Protection Agency). The screening MOE for lithium was set at 1000, equal to the total UF used in the assessment. None of the calculated MOEs were less than the screening value. These results suggest that exposure to these pharmaceuticals from drinking water is not likely to pose a public health concern. However, it is still important to note that additional toxicity data are needed and would provide greater confidence in the MOEs calculated for these pharmaceuticals.

Relatively high concentrations of lithium (compared to other pharmaceuticals) were detected in treated drinking water. Lithium was classified as a pharmaceutical in this study because its presence in source and treated waters was inferred to derive in part from lithium excreted as a result of its use as a neuroleptic pharmaceutical. Although it is not possible, based on this study, to determine whether the concentrations of lithium in source water can be apportioned between anthropogenic wastewater discharges (including pharmaceutical use) or naturally occurring lithospheric sources due to the transport of lithium from source to treated water, lithium was conservatively transported through drinking water treatment (Furlong et al., 2016). The concentrations of lithium in source and treated drinking water observed in this study are within the range of the concentrations of lithium observed in studies showing a statistically significant inverse association with suicide rates and standardized mortality rations for suicide, suggesting a potential human health effect from this exposure (Kapusta et al., 2011). Some additional data also suggest a potential for neurodevelopmental effects from prenatal exposure to lithium (Schrauzer, 2002; Gentile, 2010). None of this information was included in the health assessment for lithium (U.S. Environmental Protection Agency). Further research is necessary to decide whether controls to limit exposure to lithium are warranted to increase public health protection.

Table 1
MOE for Elemental Analytes.

Analyte	Toxicity value (mg/kg-day)	Literature reference for toxicity value	DWEL (mg/L)	Maximum detected (mg/L)	DWTP with maximum detection	MOE calculated	MOE screen
Aluminum	100 LOAEL	7	3333	0.1875	14	18000	100
Bromide	7 NOAEL	17	233.3	0.24	25	970	10
Chlorate	0.9 BMDL	15	30.00	0.32	27	94	30
Iron	1 LOAEL	7	33.33	0.0907	27	370	1.5
Manganese	0.047 NOAEL	8	1.555	0.0556	18	28	3
Nickel	5 NOAEL	8	166.7	0.0035	4	48,000	300
Silicon	800 NOAEL	18	26,670	22.26	5	1200	3000
Strontium	190 NOAEL	8	6333	0.9996	28	6300	300
Tin	32 NOAEL	14	1067	0.0159	24	67,000	100
Vanadium	0.22 NOAEL	7	7.333	0.0049	5	1500	3000
Zinc	0.91 NOAEL	8	30.33	0.1002	4	300	3

Table 2
MOE for anthropogenic waste indicators.

Analyte	Toxicity value (mg/kg-day)	Literature reference for toxicity value	DWEL (mg/L)	Maximum detected (mg/L)	DWTP with maximum detection	MOE calculated	MOE screen
Acetophenone	423 NOAEL	8	14,100	580×10^{-6}	29	24×10^{6}	3000
Hexahydrohexamethyl Cyclopentabenzopyran (HHCB, Galaxolide)	150 NOAEL	18	5000	61×10^{-6}	26	82×10^{6}	3000
Isopherone	150 NOAEL	8	5000	32×10^{-6}	2	160×10^{6}	1000
Metolachlor	9.7 NOAEL	15	323.3	100×10^{-6}	21	3.2×10^{6}	100
Triethyl citrate	2000 ADI	16	66670	13×10^{-6}	1	5100×10^{6}	100

3.4. Assessment of hormonally active environmental contaminants

Twelve hormonally active agents were included in this study and are listed in Supporting Information Table 6.

Seven hormonally active agents were detected in source water in the ng/L range. Only progesterone was detected in treated drinking water at a concentration of 0.20 ng/L in one DWTP (Glassmeyer et al., 2016). Using the LOAEL of 3.3 mg/kg body weight/day from a study (Shangold et al., 1991) used to derive the ADI for progesterone, the calculated MOE is 550 million compared to the screening MOE of 1000, which indicates that exposure to the concentration of progesterone found in this study is not likely to pose a public health concern.

In a companion effort utilizing samples collected at the same time as those reported in the present paper, Conley et al. (Conley et al., 2016) analyzed the treated drinking water from these 25 plants for three natural estrogens, estrone (E1), 17β-estradiol, (E2), estriol (E3), and one synthetic estrogen, 17α-ethinyl estradiol (EE2). These four compounds, if present in any of the treated water samples, were below the lowest concentration minimum reporting limit. In contrast, *in vitro* estrogenicity, assessed with the T47D-KBluc assay, was detected in three samples of treated drinking water. When expressed as 17β-estradiol equivalents, the maximum value detected in DWTP 1 was 0.0782 ng/L. Using the NOAEL of 5000 ng/kg-day from WHO (2000) to calculate the DWEL, the calculated MOE is 2.10 million compared to a screening MOE of 100, which indicates that exposure to the concentration of estrogenic hormones found in this study is not likely to pose a public health concern. The results from Conley et al. (2016) highlight the utility of integrated chemical and biological characterization of complex mixtures, as has been demonstrated for environmentally realistic, complex mixtures of disinfection byproducts (Simmons et al., 2008), in particular for assessing the components or fractions of the complex mixture associated with toxicity and potential risk (Rice et al., 2008).

4. Future directions

Because new chemicals and pharmaceuticals are constantly being introduced into commerce, on-going research on the presence of contaminants in drinking water is necessary. In particular it will be important to consider the relative potential human health risk(s) associated with the presence in drinking water of chemical contaminants derived from the source water along with those that may be associated with contaminants formed during disinfection (disinfection byproducts) and those that may be posed by residual microbial (bacterial, viral) contaminants. This will allow risk management and risk remediation efforts to be focused on the greatest potential risks. A potential source for new analytes to be considered for future studies is USEPA's Contaminant Candidate List (U. S. Environmental Protection Agency). Further, additional health effects data for some contaminants with limited data would help strengthen the conclusions on the public health significance from exposure to contaminants.

The analysis presented does not consider potential toxicological interactions among the CECs and other contaminants that are present in the treated drinking water from each DWTP. This type of analysis could be conducted in the future.

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Table 3
MOE for pharmaceuticals detected in treated drinking water.

Analyte	DWTP with maximum detection	Toxicity value (mg/kg-day)	Reference for toxicity value	DWEL (mg/L)	Maximum detection (ng/L)	MOE calculated
Bupropion	26	7.5 MRTD	21	250	10.9	23,000,000
Carbamazepine	23	26.7 MRTD	21	890	26.5	34,000,000
Clofibric Acid	14	33.3 MRTD	21	1110	91.7	12,000,000
Cotinine	4	None	–	–	15.8	–
Diazepam	4	0.667 MRTD	21	22.23	0.85	26,000,000
Lamivudin	17	5 MRTD	21	166.7	27.7	6,000,000
Lithium	20	2 NOAEL	7	66.67	42,700	1,600
Metoprolol	4	6.67 MRTD	21	223.3	18.4	12,000,000
Propranolol	27	10.7 MRTD	21	356.7	2.5	140,000,000
Pseudoephedrine	27	4 MRTD	21	133.3	3.75	36,000,000
Sulfamethoxazole	5	None	–	–	8.2	–
Verapamil	21	None	–	–	26.7	–

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2016.03.146.

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